

# Non-Markovianity in a non-thermal bath

Sheng-Wen Li,<sup>1</sup> Moochan B. Kim,<sup>1</sup> and Marlan O. Scully<sup>1,2,3</sup>

<sup>1</sup>*Texas A&M University, College Station, TX 77843*

<sup>2</sup>*Princeton University, Princeton, NJ 08544*

<sup>3</sup>*Baylor University, Waco, TX 76798*

(Dated: April 12, 2016)

We study the dynamics of an open quantum system interacting with a non-thermal bath. Here, “non-thermal” means that the bath modes do not need to have the same temperature, but they have an effective temperature distribution. We find that, when a quantum system is interacting with such a non-thermal bath far from thermal equilibrium, it is no longer proper to use any coarse-grained Markovian description for the system, even when their coupling strength is quite weak. Especially, when there is coherent transition with strong interference strength in the quantum system, the Markovian master equation would bring in a serious problem of negative probability. After we consider some proper non-Markovian corrections, the problem can be naturally resolved.

PACS numbers: 03.65.Yz, 05.30.-d, 05.70.Ln

## I. INTRODUCTION

The property of the bath has an critical influence to the dynamical behavior of an open quantum system. For example, common bath could give rise to decoherence-free subspace and dark state, while independent baths could not [1–4]; Squeezed baths could afford a thermal machine beyond the Carnot efficiency, which is the upper limit for canonical thermal baths [5]. These physical effects all result from the special properties of their particular baths.

When we study the dynamics of an open quantum system, we usually consider it to be interacting with a bath in thermal equilibrium, which is described by the canonical Gibbs state,

$$\rho_B = \frac{1}{Z_B} \exp\left[-\frac{1}{T} \hat{H}_B\right]. \quad (1)$$

Here  $\hat{H}_B$  is the Hamiltonian of the bath and  $T$  is the temperature (we set the Boltzmann constant  $k_B = 1$ ). Thermal equilibrium is an idealistic physics model, and non-thermal baths also exists quite widely in realistic physical world. The different bath modes in a non-thermal bath do not have to share the same temperature  $T$  as Eq. (1), but they could have an effective temperature distribution [6]. In the studies of biology systems whose environments are usually quite complicated [7–9], this non-thermal bath model could provide a description more close to the realistic situation.

In this paper, we study the dynamics of an open quantum system in such a non-thermal bath. We find that, if the bath state is far from thermal equilibrium, the relaxation of the bath will be quite slow. And it is no longer appropriated to use constant decay rates to describe the evolution of the open system. Due to the slow relaxation of the bath, the decay rates of the system also vary slowly, and sometimes they could even become negative. As the result, the dynamics of the open system shows typical non-Markovian features, and we cannot describe the system by a coarse-grained Markovian master equation as before, although the coupling strength between the system and the bath is very weak. This is different from most of previous studies where the non-Markovianity is usually caused by the strong system-bath coupling [10–15].

Especially, we are interest in the case when the quantum system has coherent transitions, also well-known as the Fano-Agarwal interference [16–18]. In this case, when the coherent transition has maximum interference, the Markovian master equation would bring in a serious negative probability problem, even when the system-bath coupling strength is quite weak. Indeed, this negative probability problem resulted from the coherent transition also exists when the bath is a thermal one, but the negative value is negligibly small and it only lasts for very short time. However, when the bath is very far from thermal equilibrium, this negative probability is significantly large and exists for very long time, which is intolerable. We find that if the non-Markovian correction is taken into consideration, this negative probability problem could be naturally resolved.

This paper is organized as follows. In Sec. II, we give a brief discussion about the property of a non-thermal bath. Then we study the time-dependent evolution of a three-level system interacting with a non-thermal bath in Sec. III, and discuss the validity of the approximations made for the master equation. We will show that there will be negative probability when the coherent transition has maximum interference. In Sec. IV, we show that this negative probability problem can be resolve by introducing non-Markovian correction. We finally draw conclusion in Sec. V.

## II. NON-THERMAL BATH

In this section, we discuss the properties of the non-thermal bath we are going to study in this paper. Usually when we study the dynamics of an open quantum system, we consider that it is coupled with a boson bath in thermal equilibrium, which is described by the thermal state

$$\rho_B = \frac{1}{Z_B} e^{-\hat{H}_B/T} = \frac{1}{Z_B} \exp\left[-\frac{1}{T} \sum_k \omega_k \hat{b}_k^\dagger \hat{b}_k\right], \quad (2)$$

where  $\hat{H}_B = \sum_k \omega_k \hat{b}_k^\dagger \hat{b}_k$  is the self Hamiltonian of the boson bath, and  $T$  is the temperature. When the temperature  $T \rightarrow 0$ ,

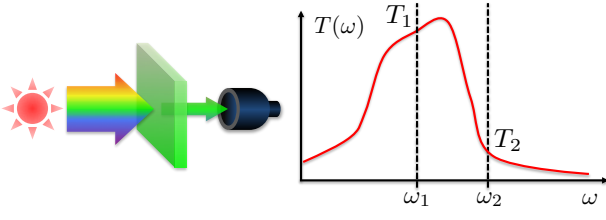


FIG. 1. (Color online) Demonstration for the effective temperature distribution of the bath modes. A filter in front of a thermal source can be utilized to modify the effective temperature distribution, and the effective temperature for each bath mode can be measured by a photon detector.

the bath tends to the vacuum state,  $\rho_B \rightarrow |\text{vac}\rangle\langle\text{vac}|$ , and there is no excitation in each boson mode.

Thermal equilibrium is an idealistic physics model, and non-thermal baths widely exist in realistic world [6, 9, 19]. We should say the thermal state is a good enough physical model within certain finite space and time. For example, a simple filter in front of a thermal light source can be utilized to create a non-thermal state (Fig. 1). The states and the energy distribution of the EM field modes on the two sides are obviously different.

Thus, we extend our requirement for the bath to be a non-thermal one, but we still assume: 1. The total bath state is a product state of each bath mode, i.e.,  $\rho_B = \rho_{k_1} \otimes \rho_{k_2} \otimes \dots$ . 2. Each mode state  $\rho_k$  has a thermal form with an effective temperature  $T_k = \beta_k^{-1}$ , i.e.,  $\rho_k \propto \exp[-\beta_k \omega_k \hat{b}_k^\dagger \hat{b}_k]$ . That is, the non-thermal state of the bath we study here is

$$\rho_B = \prod_k \frac{1}{Z_k} \exp\left[-\frac{\omega_k}{T_k} \hat{b}_k^\dagger \hat{b}_k\right]. \quad (3)$$

In this non-thermal state, the different bath modes have a distribution of effective temperatures  $\{T_k\}$ , but do not need to have a constant value  $T$ .

In principle, we can measure the state of each bath mode by optical tomography, and obtain a Wigner function description for each optical mode. For example, for the EM field under the non-thermal state (3), the expectation value of the Poynting vector is

$$\begin{aligned} \langle \hat{\mathcal{P}} \rangle &= \frac{1}{\mu_0} \langle \hat{\mathbf{E}} \times \hat{\mathbf{B}} \rangle = \sum_{\mathbf{k}, \sigma} \frac{c \hat{\mathbf{e}}_{\mathbf{k}}}{V} \cdot \hbar \omega_{\mathbf{k}} \left[ \langle \hat{b}_{\mathbf{k}\sigma}^\dagger \hat{b}_{\mathbf{k}\sigma} \rangle + \frac{1}{2} \right] \\ &= \sum_{\mathbf{k}, \sigma} \frac{c \hat{\mathbf{e}}_{\mathbf{k}}}{V} \cdot \hbar \omega_{\mathbf{k}} \bar{n}_p(\omega_{\mathbf{k}}, T_{\mathbf{k}}), \end{aligned} \quad (4)$$

where  $\hat{\mathbf{e}}_{\mathbf{k}}$  is the direction of the wave vector  $\mathbf{k}$ .  $\hat{\mathbf{E}} = -\partial_t \hat{\mathbf{A}}$ ,  $\hat{\mathbf{B}} = \nabla \times \hat{\mathbf{A}}$ , and  $\hat{\mathbf{A}}$  is the field vector,

$$\hat{\mathbf{A}} = \sum_{\mathbf{k}\sigma} \hat{\mathbf{e}}_{\mathbf{k}\sigma} \left[ \frac{\hbar}{2\varepsilon_0 \omega_{\mathbf{k}} V} \right]^{\frac{1}{2}} (\hat{b}_{\mathbf{k}\sigma} e^{i\mathbf{k}\cdot\mathbf{r} - i\omega_{\mathbf{k}} t} + \text{h.c.}). \quad (5)$$

Here we denote  $\hat{\mathbf{e}}_{\mathbf{k}\sigma}$  as the polarization direction [4, 20].

The Poynting vector  $\langle \hat{\mathcal{P}} \rangle$  describes the total energy flow passing through a unit section in unit time. Each summation term in  $\langle \hat{\mathcal{P}} \rangle$  is the photon flux of certain optical mode,

which is just the quantity measured by the photon detector. Notice that in the above Poynting vector, it contains  $\bar{n}_p(\omega, T) := [\exp(\omega/T) - 1]^{-1}$ , which is the Planck distribution. Therefore, once we measure the light intensity accepted by the photon detector, we obtain the effective temperature  $T_k$  for a certain optical mode (Fig. 1).

In the idealistic case of a thermal bath, the temperatures for each bath mode all equal to a constant value  $T_k = T$ . In more realistic cases, this requirement do not need to be fulfilled, and so we obtain a distribution  $\{T_k\}$ . If the distribution  $\{T_k\}$  is far from a constant one, we say this non-thermal bath state is far from the thermal equilibrium.

### III. DYNAMICS OF A THREE-LEVEL SYSTEM IN A NON-THERMAL BATH

#### A. Born-Markovian approximation

Now we study the dynamics of a three-level system weakly coupled with a non-thermal boson bath (Fig. 2). The Hamiltonian of the system is

$$\hat{H}_S = \varepsilon_1 |e_1\rangle\langle e_1| + \varepsilon_2 |e_2\rangle\langle e_2|, \quad (6)$$

and here we set the energy for the ground state  $|g\rangle$  to be  $\varepsilon_g = 0$ . The interaction between the system and the bath reads

$$\hat{H}_{SB} = \sum_n \hat{\tau}_n^+ \cdot \hat{B}_n + \hat{\tau}_n^- \cdot \hat{B}_n^\dagger \quad (7)$$

where  $\hat{B}_n = \sum_k g_{n,k} \hat{b}_k$  is the collective operator of the bath, and  $\hat{\tau}_n^- = |g\rangle\langle e_n|$ ,  $\hat{\tau}_n^+ = |e_n\rangle\langle g|$  are the lowering and raising operator of the 3-level system associate with the transition between  $|g\rangle$  and  $|e_n\rangle$ . Notice that here we have made the rotating-wave approximation (RWA) and omitted all the double creation/annihilation terms, because we consider that the interaction strength is quite weak and the RWA still applies.

The bath is modeled as a collection of boson modes,  $\hat{H}_B = \sum_k \omega_k \hat{b}_k^\dagger \hat{b}_k$ , as mentioned above. We need to derive a master equation to describe the dynamics of this 3-level system, which comes from the iteration of the von Neumann equation,

$$\begin{aligned} \frac{d\rho_S}{dt} &= \text{Tr}_B \left\{ -i[\rho_{SB}(t), \hat{H}_{SB}(t)] \right. \\ &\quad \left. - \int_0^t ds [\rho_{SB}(s), \hat{H}_{SB}(s)], \hat{H}_{SB}(t)] \right\}. \end{aligned} \quad (8)$$

This is an exact equation in the interaction picture of  $\hat{H}_S + \hat{H}_B$ , but still not easy for practical calculations.

We still need some assumptions to simplify the above equation. The first one is the Born approximation, i.e., during the evolution, the total state of the open system and the bath is  $\rho_{SB}(t) \simeq \rho_S(t) \otimes \rho_B(0)$ . Namely, we consider the bath is so large that the system almost cannot change bath state. Besides, the relaxation time of the bath  $\tau_B$  is usually much shorter than the decay time of the open system  $\tau_S$ , so the bath could “refresh” to its original state quickly before the

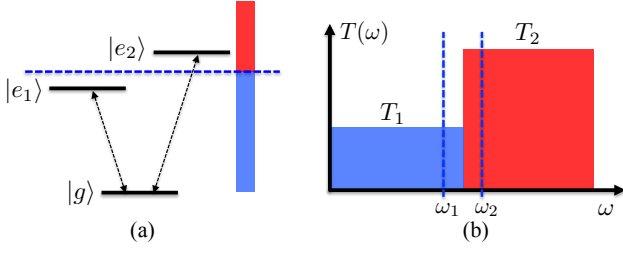


FIG. 2. (Color online) Demonstration for (a) the three-level atom and its transition structure, (b) the effective temperature distribution of the bath modes.

open system evolves [11]. Here, although the bath is a non-thermal state [Eq. (3)], we still assume the Born approximation is valid, so the above equation becomes

$$\begin{aligned} \frac{d\rho_S}{dt} &\simeq -\text{Tr}_B \int_0^t ds [\rho_S(s) \otimes \rho_B, \hat{H}_{SB}(s)], \hat{H}_{SB}(t) \quad (9) \\ &= -\text{Tr}_B \int_0^t ds' [\rho_S(t-s') \otimes \rho_B, \hat{H}_{SB}(t-s')], \hat{H}_{SB}(t). \end{aligned}$$

Notice that the first term of Eq. (8) vanishes, because for the non-thermal state Eq. (3), we always have  $\langle \hat{B}_n(t) \rangle = \text{Tr}_B[\rho_B \cdot \sum g_{n,k} \hat{a}_k \exp(-i\omega_k t)] = 0$ .

Now we obtain a differential-integral equation for  $\rho_S(t)$ . It contains a time-nonlocal convolution term, which means, if we want to know the changing rate of  $\rho_S(t)$ , we need to accumulate all the information of  $\rho_S(s')$  for  $0 \leq s' \leq t$ , not only the instantaneous value of  $\rho_S$  at  $t$ .

At this stage, we further assume that the convolution kernel in the integral, which comes from the time correlation function of the bath operators, decays so fast with  $t-s'$  that only the accumulation around  $\rho(t-s' \simeq t)$  dominates in the integral. Thus, from Eq. (9) we obtain

$$\frac{d\rho_S}{dt} \simeq -\text{Tr}_B \int_0^t ds' [\rho_S(t) \otimes \rho_B, \hat{H}_{SB}(t-s')], \hat{H}_{SB}(t). \quad (10)$$

This is a differential equation local in time, and here we call this approximation *Markov-1*. If we use the cumulant expansion method of van Kampen, also known as the time-convolutionless (TCL) method, to derive a non-Markovian master equation, Eq. (10) is just its lowest order (TCL-2) [11].

Usually we further extend the integral to infinity approximately, because we assumed that the time correlation functions of the bath approach to their steady values quite fast, and here we call it *Markov-2*. After the approximation Markov-2, we obtain

$$\frac{d\rho_S}{dt} \simeq -\text{Tr}_B \int_0^\infty ds' [\rho_S(t) \otimes \rho_B, \hat{H}_{SB}(t-s')], \hat{H}_{SB}(t). \quad (11)$$

This is usually the starting point for the derivation of the Markovian master equation.

The Liouville operator obtained from Markov-2 [Eq. (11)] usually does not depend on time explicitly (in Schrödinger's

picture) [21, 22], which brings us great convenience for calculation, but we should keep in mind that Markov-2 indeed ignored the precise dynamical behaviour within the bath correlation time  $\tau_B$ . That is, for the period  $0 \leq t \lesssim \tau_B$ , the approximation

$$\int_0^t ds' \dots \simeq \int_0^\infty ds' \dots \quad (12)$$

is indeed not quite reliable, but we just do not care about the dynamics within this short time.

## B. Coherent transition and secular approximation

Starting from Markov-2 [Eq. (11)], we can obtain the following master equation in Schrödinger's picture (the details of the derivation is shown in Appendix A, see also Ref. [22]),

$$\begin{aligned} \dot{\rho}_S &= i[\rho_S, \hat{H}_S + \hat{H}_c] \\ &+ \sum_{m,n=1}^2 D_{mn}^+ (\hat{\tau}_m^+ \rho_S \hat{\tau}_n^- - \frac{1}{2} \{ \hat{\tau}_n^- \hat{\tau}_m^+, \rho_S \}_+) \\ &+ D_{mn}^- (\hat{\tau}_m^- \rho_S \hat{\tau}_n^+ - \frac{1}{2} \{ \hat{\tau}_n^+ \hat{\tau}_m^-, \rho_S \}_+), \quad (13) \end{aligned}$$

where we denote the decay rates as  $D_{mn}^\pm := [\Gamma_{mn}^\pm(\varepsilon_m) + \Gamma_{mn}^\pm(\varepsilon_n)]/2$ , and

$$\begin{aligned} \Gamma_{mn}^+(\omega) &:= J_{nm}(\omega) \tilde{n}(\omega), \\ \Gamma_{mn}^-(\omega) &:= J_{mn}(\omega) [\tilde{n}(\omega) + 1]. \quad (14) \end{aligned}$$

$J_{mn}(\omega)$  is the coupling spectrum, which is defined by

$$J_{mn}(\omega) := 2\pi \sum_k g_{m,k}^* g_{n,k} \delta(\omega - \omega_k) = [J_{nm}(\omega)]^*. \quad (15)$$

$\tilde{n}(\omega)$  is the photon number distribution obtained from  $\tilde{n}(\omega_k) = \text{Tr}[\rho_B \cdot \hat{a}_k^\dagger \hat{a}_k]$ . For a non-thermal state like Eq. (3), we have

$$\tilde{n}(\omega) = \frac{1}{\exp \frac{\omega}{T(\omega)} - 1}, \quad (16)$$

where  $T(\omega)$  is the effective temperature distribution. If  $T(\omega)$  is a constant distribution  $T(\omega) = T$ ,  $\tilde{n}(\omega)$  could return to the Planck distribution  $\bar{n}_p(\omega, T)$  for the thermal bath.

Notice that except the dissipation part in the master equation, we also have a correction in the unitary term [22],

$$\begin{aligned} \hat{H}_c &:= \sum_{m,n=1}^2 \frac{1}{4i} [\Gamma_{mn}^+(\varepsilon_m) - \Gamma_{mn}^+(\varepsilon_n)] \cdot \hat{\tau}_n^- \hat{\tau}_m^+ \\ &+ \frac{1}{4i} [\Gamma_{mn}^-(\varepsilon_m) - \Gamma_{mn}^-(\varepsilon_n)] \cdot \hat{\tau}_n^+ \hat{\tau}_m^-, \quad (17) \end{aligned}$$

and  $\hat{H}_c$  is not from the principal integral. Here we omitted the principal integrals in the above master equation.

From the definition of  $J_{mn}(\omega)$  [Eq. (15)], we directly obtain the property  $J_{mn}(\omega) = [J_{nm}(\omega)]^*$ , and this guarantees  $\hat{H}_c = \hat{H}_c^\dagger$ . Here we have both individual spectrums  $J_{nn}(\omega)$  and cross spectrums  $J_{mn}(\omega)$  for  $m \neq n$  [22, 23]. The individual spectrum  $J_{nn}(\omega)$  corresponds to the decay/excitation behavior of each transition path itself, while the cross spectrum  $J_{mn}(\omega)$  ( $m \neq n$ ) corresponds to the coherent interference between different transition paths. This effect is often called the Fano-Agarwal interference [16]. It can be proved that we always have  $|J_{mn}(\omega)|^2 \leq J_{mm}(\omega) \cdot J_{nn}(\omega)$  [22]. Thus we define a coherence strength  $p(\omega)$ , so that  $J_{12}(\omega) = p(\omega) \cdot \sqrt{J_{11}(\omega)J_{22}(\omega)}$ , to measure the interference strength of the coherent transition. When  $|p(\omega)| = 1$ , we have the maximum interference effect, while  $p(\omega) = 0$  indicates there is no interference between the two transitions. For a specific physics model, this coherence strength  $p(\omega)$  can be calculated explicitly from the system-bath interaction.

It should be notice that, in the interaction picture, these coherent transition terms have time-dependent coefficients  $\exp[\pm i\Delta_{mn}t]$  when the two levels are not degenerated  $\Delta_{mn} := \varepsilon_m - \varepsilon_n \neq 0$ . Thus, these terms are often dropped by the secular approximation in order to directly get a Lindblad form with explicitly positive decay rates [8, 11, 24]. However, this secular approximation indeed implies that the precise evolution details within the time scale  $\tau_\Delta \simeq \hbar/\Delta_{mn}$  is omitted. When  $\Delta_{mn}$  is a small value,  $\tau_\Delta$  is comparable with the system decay time  $\tau_S$ , and thus this is not a good enough approximation. Besides, from the above discussion of the cross spectrum  $J_{mn}(\omega)$ , we see that these coherent transition terms do have a clear physical meaning, and they play an essential role in some physical effects like coherent population trapping and dark state, which has been well known both theoretically and experimentally [4, 16, 17]. Moreover, it was also found that these coherent transitions are closely connected with the non-equilibrium flux in some transport systems, and if they are omitted, we will obtain some unphysical results [21, 22, 24]. Therefore, the secular approximation should not be made at this stage.

In sum, in the derivation of the master equation (13), we used the Born approximation, Markov-1 and Markov-2. Usually when we talk Markovian approximation in literature, we mean Markov-1 and Markov-2 together. Now we are going to show that for the non-thermal bath case, we need to separate them apart, and indeed Markov-2 is not a good enough approximation. More importantly, we should keep in mind that the name ‘‘Markovian approximation’’ never promises us to give a legitimate Markovian dynamical behavior, as we will show below.

### C. The problem of negative probability

Now we can use the master equation (13) to study the dynamics of the 3-level system. The influence of the non-thermal bath state is contained in the modified photon number distribution  $\tilde{n}(\omega)$ . For simplicity, we consider a distribution

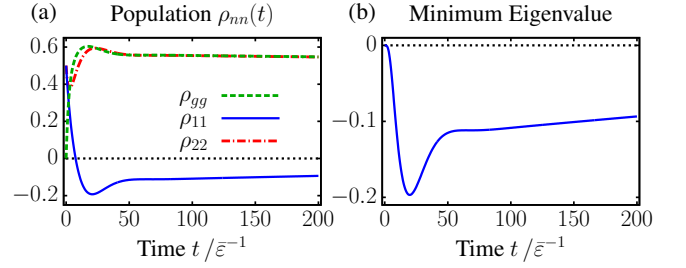


FIG. 3. (Color online) Time-dependent evolution of  $\rho_S(t)$  in a non-thermal bath calculated from the master equation (13). (a) The populations  $\rho_{nn}(t)$  and (b) The smallest eigenvalue of  $\rho_S(t)$ .  $\rho_{11}(t) := \langle e_1 | \rho_S(t) | e_1 \rangle$  become negative. Here we set  $\bar{\varepsilon} := (\varepsilon_1 + \varepsilon_2)/2 \equiv 1$  as the unit, and  $\varepsilon_1 = 0.95$ ,  $\varepsilon_2 = 1.05$ . The interference strength is  $p = 1$ . The decay rate is  $J_{mn}(\varepsilon_{1,2}) \equiv \gamma = 0.0005$  for  $m, n = 1, 2$ . The two effective temperatures are  $T_1 = 1$ ,  $T_2 = 500$ , which is very far from the thermal equilibrium.

as follows [Fig. 2(b)],

$$T(\omega) = \begin{cases} T_1, & 0 < \omega \leq \bar{\varepsilon}, \\ T_2, & \omega > \bar{\varepsilon}, \end{cases} \quad (18)$$

where  $\bar{\varepsilon} := (\varepsilon_1 + \varepsilon_2)/2$  just lies in the middle between the two energy gap of the 3-level system.

Due to the change of the photon number distribution, the decay rates in the master equation does not satisfy the detailed balance or micro-reversibility [25, 26]. Intuitively, if we do not consider the coherent transition induced by the cross coupling spectrum, we can regard the two excited state  $|e_1\rangle$  and  $|e_2\rangle$  as two independent systems immersed in two thermal baths with temperatures  $T_1$  and  $T_2$  respectively.

We calculate the time-dependent evolution of the 3-level system in a non-thermal bath according to the master equation (13), and we show the result in Fig. 3. Strikingly, if the evolutions start from the initial state  $|\psi_0\rangle = (|e_1\rangle + |e_2\rangle)/\sqrt{2}$ , the population  $\rho_{22}(t) := \langle e_2 | \rho_S(t) | e_2 \rangle$  on the energy level  $|e_2\rangle$  becomes negative, and this negative probability lasts for a very long time (Fig. 3) [7, 8].

The diagonal terms  $\rho_{nn}(t)$  of the density matrix means the probabilities that the system stays in this state  $|n\rangle$  at time  $t$ , and they should never be negative. The density matrix must be semi-positive at any time. Indeed this negative probability problem also exists when the bath is a thermal state. We show the evolution of the 3-level system when the bath is a thermal one (Fig. 4). In this case, the populations in each eigen energy level are always positive during the evolution starting from  $|\psi_0\rangle = (|e_1\rangle + |e_2\rangle)/\sqrt{2}$  as before. But if we further check the minimum eigenvalue of  $\rho_S(t)$ , we find that the smallest eigenvalue of  $\rho_S(t)$  becomes a negative value right after the evolution begins, and quickly turns to be positive after a very short time. That means,  $\rho_S(t)$  also has a negative probability in a certain basis even when the bath is a thermal state.

For the thermal bath case, this negative probability problem is not too serious, since the negative value is quite small, and it only lasts for a very short time (Fig. 4). In the sense of the coarse-graining idea of Markovian approximation, this



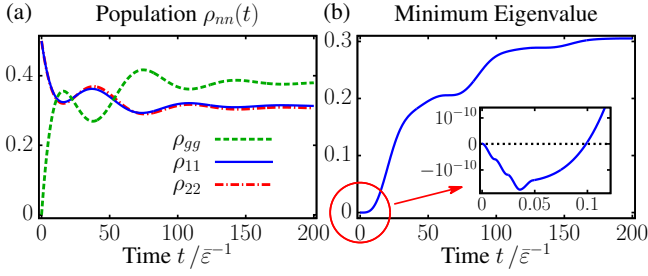


FIG. 4. (Color online) Time-dependent evolution of  $\rho_S(t)$  in a thermal bath calculated from the master equation (13). (a) The populations  $\rho_{nn}(t)$  (b) The smallest eigenvalue of  $\rho_S(t)$ . The populations in each energy levels are positive, but the minimum eigenvalue of  $\rho_S(t)$  becomes a small negative value immediately after the evolution begins, and lasts for a very short time. Here we set  $\bar{\varepsilon} := (\varepsilon_1 + \varepsilon_2)/2 \equiv 1$  as the unit, and  $\varepsilon_1 = 0.95$ ,  $\varepsilon_2 = 1.05$ . The interference strength is  $p = 1$ . The decay rate is  $J_{mn}(\varepsilon_{1,2}) \equiv \gamma = 0.01$  for  $m, n = 1, 2$ . The temperature of the thermal bath is  $T = 5$ .

inaccuracy is negligible [27]. But this problem is intolerable when the bath is a non-thermal state far from equilibrium, because the negative value is significantly large and lasts for too long time (Fig. 3).

To find the reason for this negative probability problem, we need to check the positivity condition of the master equation (13). It was proved that for a master equation like

$$\begin{aligned} \dot{\rho} &= \mathcal{L}[\rho] \\ &= i[\rho, \hat{H}] + \sum_{m,n} \kappa_{mn} (\hat{L}_m \rho \hat{L}_n^\dagger - \frac{1}{2} \{ \hat{L}_n^\dagger \hat{L}_m, \rho \}_+), \end{aligned} \quad (19)$$

starting from any legitimate density matrix  $\rho_0$ ,  $\rho(t)$  is always semi-positive at any time  $t$ , if and only if the coefficients  $\kappa_{mn}$  form a non-negative Hermitian matrix [28, 29]. This equation is called the Lindblad form or GKSL form (Gorini-Kossakowski-Sudarshan-Lindblad). Here  $\hat{H}$  is not required to be the self Hamiltonian of the system, and it is only required that  $\hat{H}$  should be a Hermitian operator.

The master equation (13) has already been written in a Lindblad form, but we need to check the positivity of the decay rate matrix  $\mathbf{D}$  for the Lindblad operators  $\{\hat{\tau}_1^+, \hat{\tau}_2^+, \hat{\tau}_1^-, \hat{\tau}_2^-\}$ , i.e.,

$$\begin{aligned} \mathbf{D} &= \begin{bmatrix} \mathbf{D}^+ & \mathbf{0} \\ \mathbf{0} & \mathbf{D}^- \end{bmatrix}, \\ \mathbf{D}^\pm &= \begin{bmatrix} \Gamma_{11}^\pm(\varepsilon_1) & \frac{1}{2}[\Gamma_{12}^\pm(\varepsilon_1) + \Gamma_{12}^\pm(\varepsilon_2)] \\ \frac{1}{2}[\Gamma_{12}^\pm(\varepsilon_1) + \Gamma_{12}^\pm(\varepsilon_2)] & \Gamma_{22}^\pm(\varepsilon_2) \end{bmatrix}, \end{aligned} \quad (20)$$

and the definition of  $\Gamma_{mn}^\pm(\omega)$  follows from Eq. (14). To prove the positivity of  $\mathbf{D}$ , it suffices to prove the positivity of the two block matrices  $\mathbf{D}^\pm$ . It is obvious we already have  $\Gamma_{nn}^\pm(\omega) \geq 0$ , thus we just need to check whether  $\det \mathbf{D}^\pm \geq 0$ , i.e.,

$$\det \mathbf{D}^\pm = \Gamma_{11}^\pm(\varepsilon_1) \Gamma_{22}^\pm(\varepsilon_2) - \frac{1}{4} |\Gamma_{12}^\pm(\varepsilon_1) + \Gamma_{12}^\pm(\varepsilon_2)|^2. \quad (21)$$

In the above two examples, we assumed  $J_{nn}(\varepsilon_n) = \gamma_n$ , and  $J_{12}(\varepsilon_1) = J_{12}(\varepsilon_2) = p \cdot \sqrt{\gamma_1 \gamma_2}$ ,  $0 \leq p \leq 1$ , thus the determinants are

$$\begin{aligned} \det \mathbf{D}^+ &= (1 - p^2) \gamma_1 \tilde{n}(\varepsilon_1) \cdot \gamma_2 \tilde{n}(\varepsilon_2) \\ &\quad - \frac{p^2}{4} \cdot \gamma_1 \gamma_2 [\tilde{n}(\varepsilon_1) - \tilde{n}(\varepsilon_2)]^2, \\ \det \mathbf{D}^- &= (1 - p^2) \gamma_1 [\tilde{n}(\varepsilon_1) + 1] \cdot \gamma_2 [\tilde{n}(\varepsilon_2) + 1] \\ &\quad - \frac{p^2}{4} \cdot \gamma_1 \gamma_2 [\tilde{n}(\varepsilon_1) - \tilde{n}(\varepsilon_2)]^2. \end{aligned} \quad (22)$$

Therefore, if the coherent transition achieves the maximum interference  $p = 1$ , we always have  $\det \mathbf{D}^\pm \leq 0$ , and the equality holds if and only if  $\tilde{n}(\varepsilon_1) = \tilde{n}(\varepsilon_2)$ . That means, even for the case of a thermal bath  $\tilde{n}(\varepsilon_n) = \bar{n}_p(\varepsilon_n, T)$ , the decay matrix  $\mathbf{D}$  always contains negative eigenvalues unless  $\varepsilon_1 = \varepsilon_2$  or  $T = 0$ .

The negative eigenvalue of the decay matrix  $\mathbf{D}$  implies some modes of the system have negative decay rates. This is also why the problem of negative probability appears in the master equation (13) after the approximation Markov-2. For a thermal bath state, the difference of  $\bar{n}_p(\varepsilon_1, T)$  and  $\bar{n}_p(\varepsilon_2, T)$  are quite small when  $\Delta_{12}/T \ll 1$ , thus  $\det \mathbf{D}^\pm$  are very small although negative, and so do the negative eigenvalues of  $\mathbf{D}$ . But when the bath is a non-thermal state, the difference of  $\tilde{n}(\varepsilon_1)$  and  $\tilde{n}(\varepsilon_2)$  can be very large, and that gives rise to very large negative diverging rate. Therefore, the problem of negative probability is much more serious in a non-thermal bath as mentioned in Fig. 3.

From the determinants Eq. (22) we also notice that the decay rate matrix  $\mathbf{D}$  is always positive if the coherence strength is lower than an upper bound,

$$p \leq p_c := \frac{4\tilde{n}(\varepsilon_1)\tilde{n}(\varepsilon_2)}{[\tilde{n}(\varepsilon_1) + \tilde{n}(\varepsilon_2)]^2} \leq 1. \quad (23)$$

And the larger the difference  $|\tilde{n}(\varepsilon_1) - \tilde{n}(\varepsilon_2)|$  is, the smaller  $p_c$  we obtain. This bound is also the limit for the validity of the Markovian master equation with coherent transition, namely, if the strength of the coherent transition is greater than this bound  $p_c$ , it is no longer possible to describe the system dynamics by a homogenous Markovian master equation like Eq. (13).

#### IV. NON-MARKOVIAN DYNAMICS

We have seen that if the interference strength of the coherent transition is greater than the upper limit  $p_c$  [Eq. (23)], the positivity of the decay rate matrix  $\mathbf{D}$  in the homogeneous Markovian master equation (13) is broken down, and that brings in the problem of negative probability. This problem is intolerable when the bath state is far from thermal equilibrium because the negative probability has a significantly large value and lasts for a quite long time (Fig. 3).

However, the interference strength  $p(\omega)$  of the coherent transition is determined by the properties of the physical system itself, and there is no physical law forbidding it to achieve

its maximum value. Thus, now we need to answer, if a system with maximum coherent transition is immersed in a non-thermal bath, how to resolve this negative probability problem. Here we show that this problem can be cured by considering the time-dependence of the decay rates, no matter whether the bath is thermal or non-thermal. And we will see that this gives rise to non-Markovian dynamics.

### A. Correlation time

First, we release the approximation Markov-2 [Eq. (11)], and derive another time-dependent master equation based on Markov-1 [Eq. (10)], which reads (see derivations in Appendix A),

$$\begin{aligned} \dot{\rho}_S = & i[\rho_S, \hat{H}_S + \hat{H}_c(t)] \\ & + \sum_{m,n=1}^2 D_{mn}^+(t) (\hat{\tau}_m^+ \rho_S \hat{\tau}_n^- - \frac{1}{2} \{ \hat{\tau}_n^- \hat{\tau}_m^+, \rho_S \}_+) \\ & + D_{mn}^-(t) (\hat{\tau}_m^- \rho_S \hat{\tau}_n^+ - \frac{1}{2} \{ \hat{\tau}_n^+ \hat{\tau}_m^-, \rho_S \}_+). \end{aligned} \quad (24)$$

Comparing with the homogeneous Markovian master equation (13), the decay rates become time-dependent now, i.e.,  $D_{mn}^\pm(t) := [\Gamma_{mn}^\pm(\varepsilon_m, t) + \Gamma_{mn}^\pm(\varepsilon_n, t)]/2$ , and

$$\begin{aligned} \Gamma_{mn}^+(\omega, t) &:= \Re \int_0^t ds \int_0^\infty \frac{d\nu}{2\pi} J_{mn}(\nu) \tilde{n}(\nu) e^{i(\nu-\omega)s} \\ \Gamma_{mn}^-(\omega, t) &:= \Re \int_0^t ds \int_0^\infty \frac{d\nu}{2\pi} J_{mn}(\nu) [\tilde{n}(\nu) + 1] e^{i(\omega-\nu)s}. \end{aligned} \quad (25)$$

Notice that  $\Gamma_{mn}^-(\omega, t) = \Gamma_{nm}^+(\omega, t) + \Gamma_{mn}^0(\omega, t)$ , where

$$\Gamma_{mn}^0(\omega, t) := \Re \int_0^t ds \int_0^\infty \frac{d\nu}{2\pi} J_{mn}(\nu) e^{i(\nu-\omega)s} \quad (26)$$

does not depend on the bath state. The unitary correction term  $\hat{H}_c(t)$  is the same as Eq. (17) except the corresponding changes of  $\Gamma_{mn}^\pm(\varepsilon_i, t)$ . When  $t \rightarrow \infty$ ,  $\Gamma_{mn}^\pm(\omega, t)$  could return to Eq. (14) exactly. The imaginary part of the above integrals contributes to a time-dependent Lamb shift which we do not consider here.

When the time dependence of the decay rates are considered, we need to choose a specific coupling spectrum  $J_{mn}(\omega)$  [Eq. (15)]. Here we use the linear spectrum  $J_{mn}(\omega) = \lambda_{mn}\omega$  for  $m, n = 1, 2$  (also known as the Ohmic spectrum), with a step function  $\Theta(\Omega_c - \omega)$  as the cutoff. When the cutoff  $\Omega_c \rightarrow \infty$ , this linear coupling spectrum could lead to white noise, namely, the noise spectrum of the bath tends to be flat [11, 15, 30], which fits the idea of Markovian approximation more closely. For simplicity, we choose  $\lambda_{mn} = \lambda$  for  $m, n = 1, 2$ , which also implies the interference of the coherent transition achieves the maximum  $p = 1$ . Accordingly,  $\Gamma_{mn}^\pm(\omega, t)$  equal to each other for  $m, n = 1, 2$ , and we denote as  $\Gamma_{mn}^\pm(\omega, t) := \Gamma^\pm(\omega, t)$ .

We still start from the same initial state  $|\psi_0\rangle = (|e_1\rangle + |e_2\rangle)/\sqrt{2}$  as before, and calculate the evolution of  $\rho_S(t)$  by

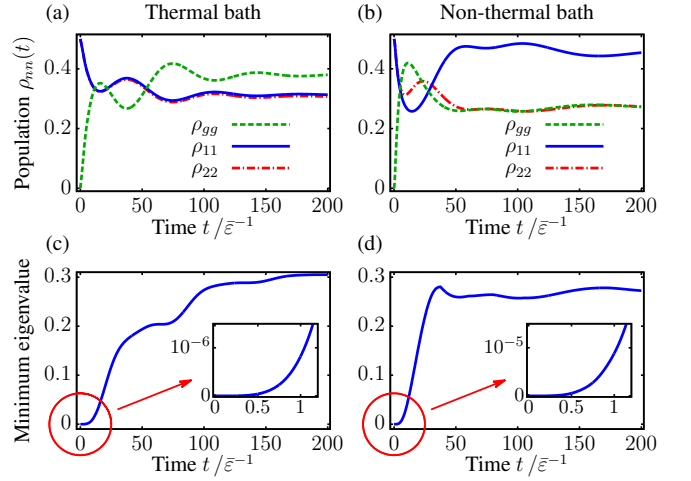


FIG. 5. (Color online) (a, b) Evolution of the populations  $\rho_{nn}(t)$  in thermal/non-thermal bath calculated from the time-dependent master equation (24). (c, d) The minimum eigenvalue of  $\rho_S(t)$  bath also keeps non-negative in thermal/non-thermal. The coupling spectrums are  $J_{mn}(\omega) = \lambda\omega \cdot \Theta(\Omega_c - \omega)$ . Here we set  $\bar{\varepsilon} := (\varepsilon_1 + \varepsilon_2)/2 \equiv 1$  as the unit, and  $\varepsilon_1 = 0.95, \varepsilon_2 = 1.05$ . The interference strength is  $p = 1$ . We choose the cutoff as  $\Omega_c = 20$ . For the thermal case (a, c), we set  $T = 5$  and  $\lambda = 0.01$  (see also Fig. 4). For the non-thermal case (b, d) we set  $T_1 = 1, T_2 = 500$  and  $\lambda = 0.0005$  (see also Fig. 3).

this time-dependent master equation (24) under the same parameters as those in Figs. 3, 4 correspondingly. We find that, no matter whether the bath is thermal or non-thermal, the populations  $\rho_{nn}(t)$  are always positive, and so does the minimum eigenvalue of  $\rho_S(t)$  (Fig. 5). The previous negative probability problem is resolved.

This result implies that the application of Markov-2 is not appropriate, which leads to the emergence of the negative probability problem as the result. The reason is, when we do the Markovian approximations as discussed in Sec. III A, we have assumed that the system decay time  $\tau_S$  is much shorter than the bath relaxation time  $\tau_B$ , but indeed this assumption is still waiting for a self-consistency examination after we obtain the master equation.

Now we do this consistency examination. We calculate the time dependence of the decay rates  $\Gamma^+(\varepsilon_{1,2}, t)$  and show them in Fig. 6(a, b). For the thermal bath,  $\Gamma^+(\varepsilon_{1,2}, t)$  quickly reach their steady values after a very short time, and then only have small oscillations around it. Therefore, it is a good enough approximation to replace  $\Gamma^+(\varepsilon_{1,2}, t)$  by their constant steady values. This is just what is done in the Markov-2 approximation.

We also see that this replacement by a constant is not good for  $\Gamma^+(\varepsilon_{1,2}, t)$  in the beginning short period. As the result, the dynamics calculated from Markov-2 is not reliable within this period even for the case of a thermal bath [see also discussion around Eq. (12)]. This is also why we have negative probability in the beginning period shown in Fig. 4. In Markovian approximations, this short time inaccuracy is omitted based on the idea of coarse-graining, and we do notice that there

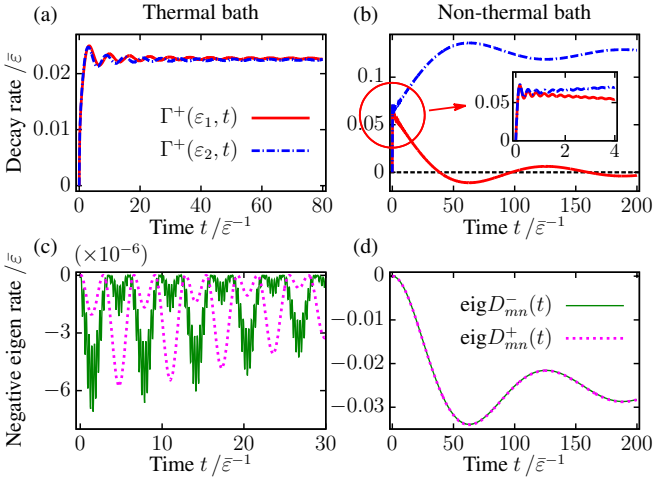


FIG. 6. (Color online) (a, b) Time-dependent decay rates  $\Gamma^+(\varepsilon_{1,2}, t)$  in thermal/non-thermal bath. (c, d) Evolution of the negative eigenvalue of the decay matrix  $D^\pm(t)$  in thermal/non-thermal bath. The coupling spectrums are  $J_{mn}(\omega) = \lambda\omega \cdot \Theta(\Omega_c - \omega)$ . Here we set  $\bar{\varepsilon} := (\varepsilon_1 + \varepsilon_2)/2 \equiv 1$  as the unit, and  $\varepsilon_1 = 0.95$ ,  $\varepsilon_2 = 1.05$ . The interference strength is  $p = 1$ . We choose the cutoff as  $\Omega_c = 20$ . For the thermal case (a, c), we set  $T = 5$  and  $\lambda = 0.01$  (see also Fig. 4). For the non-thermal case (b, d) we set  $T_1 = 1$ ,  $T_2 = 500$  and  $\lambda = 0.0005$  (see also Fig. 3). The two lines in (d) are almost overlapped.

is almost no difference between Fig. 4(a) and Fig. 5(a) even quantitatively.

For the case of a non-thermal bath far from equilibrium, the evolution of  $\Gamma^+(\varepsilon_{1,2}, t)$  can be also divided into two stages. However, after the first rapid relaxation stage, the decay rates  $\Gamma^+(\varepsilon_{1,2}, t)$  still vary significantly and slowly before they approach the steady values due to the non-thermal temperature distribution. The decay rate  $\Gamma^+(\varepsilon_1, t)$  even becomes negative at some time. Obviously we cannot replace them simply by a constant as the case of thermal bath.

We also notice that  $\Gamma^+(\varepsilon_2, t)$  tends to a large value. This is because the frequency  $\varepsilon_2$  is resonant with a bath mode with a very high effective temperature ( $T_2 = 500$ ). We should remember that the relaxation time  $\tau_S$  of the open system is characterized by  $\Gamma^\pm(\varepsilon_{1,2}, t)$ . Thus, the system relaxation time  $\tau_S$  becomes much shorter. All these facts imply that the previous basis ansatz for Markovian approximation,  $\tau_B \ll \tau_S$ , no longer holds for a non-thermal bath far from equilibrium, and the dynamics of the open system must be non-Markovian, even if the coupling strength between the system and the bath is very weak ( $\lambda = 0.0005$ ).

### B. Non-Markovianity

Besides the comparison of the correlation time as above, we have some more sufficient evidences to determine the non-Markovianity of the open quantum system. The first simple non-Markovian feature is reflected in the colored noise spectrum of the bath. The noise spectrum measured by the detector

is calculated from the Fourier transform of the symmetrized time correlation function of the bath operators [11, 30, 31], i.e.,

$$\begin{aligned} \bar{S}_{mn}(\omega) &:= \frac{1}{2} \int_{-\infty}^{\infty} ds \text{Tr} \left[ \rho_B \cdot \{ \hat{B}_m^+(t), \hat{B}_n^-(t+s) \}_+ \right] e^{i\omega s} \\ &= J_{mn}(\omega) [\tilde{n}(\omega) + \frac{1}{2}]. \end{aligned} \quad (27)$$

For the thermal bath case, we have  $\tilde{n}(\omega) = \bar{n}_p(\omega, T)$ . If we use the linear spectrum  $J_{mn}(\omega) = \lambda_{mn}\omega$ , with the help of the expansion,

$$\omega \left[ \frac{1}{e^{\beta\omega} - 1} + \frac{1}{2} \right] \simeq \frac{1}{\beta} + \frac{\beta\omega^2}{12} + \dots \quad (28)$$

we obtain a flat noise spectrum  $\bar{S}_{mn}(\omega) \simeq \lambda_{mn}T$  at high temperature limit,  $\beta = T^{-1} \rightarrow 0$ , i.e.,  $\bar{S}_{mn}(\omega)$  is a constant and does not depend on  $\omega$ . This is just the white noise spectrum which leads to Markovian dynamics [11, 31]. However, if the bath has a non-trivial temperature distribution [e.g., like Eq. (18)], the noise spectrum would become  $\bar{S}_{mn}(\omega) \simeq \lambda_{mn}T(\omega)$ , which is a colored noise spectrum. That implies the emergence of non-Markovian dynamics.

There are many different approaches to determine the non-Markovianity of a quantum process [32]. Based on the divisibility criterion of the dynamical map, it was proved that a master equation like

$$\begin{aligned} \dot{\rho} &= \mathcal{L}_{(t)}[\rho] \\ &= i[\rho, \hat{H}(t)] + \sum_{m,n} \kappa_{mn}(t) \left( \hat{L}_m \rho \hat{L}_n^\dagger - \frac{1}{2} \{ \hat{L}_n^\dagger \hat{L}_m, \rho \}_+ \right) \end{aligned} \quad (29)$$

gives a quantum Markovian process, if and only if the decay matrix  $[\kappa_{mn}(t)]$  is semi-positive at any time  $t$  [32, 33].

Thus, we check the positivity of the decay matrix of the time-dependent master equation (24). When we have maximum interference  $p = 1$  and  $\Gamma_{mn}^\pm(\varepsilon_{1,2}, t) = \Gamma^\pm(\varepsilon_{1,2}, t)$  as above, we can prove that the determinants of the block matrices,

$$D^\pm(t) = \begin{bmatrix} \Gamma^\pm(\varepsilon_1, t) & \frac{1}{2} [\Gamma^\pm(\varepsilon_1, t) + \Gamma^\pm(\varepsilon_2, t)] \\ * & \Gamma^\pm(\varepsilon_2, t) \end{bmatrix}, \quad (30)$$

in the decay matrix  $D(t)$  are always negative,  $\det D^\pm(t) \leq 0$ . That means  $D(t)$  always have one positive and one negative eigenvalue. We show the evolution of these two negative eigenvalues in Fig. 6(c, d) for both thermal/non-thermal bath cases. These eigenvalues are negligibly small in the thermal bath case, while significantly large in the non-thermal bath. For the non-thermal bath case, even the diagonal terms of  $D(t)$  themselves,  $\Gamma^\pm(\varepsilon_{1,2}, t)$ , could also become negative [Fig. 6(b)]. Therefore, even when there is no coherent transition,  $p = 0$ , it is still necessary to consider the non-Markovian effect in a non-thermal bath. As we mentioned before, “Markovian approximation” did not promise to give a Markovian process.

Therefore, we conclude that the dynamics of the open quantum system shows typical non-Markovian feature even when the system-bath coupling is very weak. It is worth noticing that in previous literatures non-Markovian dynamics is usually resulted from the ultra-strong coupling between the system and its environment [10–15]. Different from previous studies, the non-Markovianity in our study roots from the coherent transition and the non-thermal property of the bath. Especially, the non-Markovianity is greatly enhanced by the non-thermality of the bath.

Here we need to emphasize that the above criterion for Markovian process did not include how to determine whether the time-dependent master equation (29) is complete positive, and this is still an open question [34]. Indeed, if we keep increasing the system-bath coupling strength  $\lambda$  or the temperature difference  $T_2 - T_1$  in the above examples, the time-dependent master equation (24) will also have the negative problem. In that case, non-Markovian corrections of higher orders are needed.

## V. CONCLUSION

In this paper, we studied the dynamics of an open quantum system interacting with a non-thermal bath. We find that when the bath state is far from thermal equilibrium, the dynamics of the open quantum system becomes non-Markovian.

The noise spectrum of the non-thermal bath is not the flat one as the white noise case. As a result, the correlation functions of the non-thermal bath vary significantly for a long time during relaxation. These behaviors are no longer consistent with the Markovian master equation which is widely adopted in previous studies.

The coherent transition in the open quantum system could bring in the negative probability problem. For the thermal bath case, this problem is negligible in the sense of coarse-graining idea of Markovian approximation. But it is intolerable in a non-thermal bath, where the negative probability is large and exists for very long time. This negative probability problem can be naturally resolved after we consider non-Markovian corrections.

Different from previous studies, here even when the system is weakly coupled with the bath, we still cannot find a coarse-grained Markovian description for the open quantum system in a non-thermal bath. the non-Markovianity in our study is resulted from the coherent transition and the non-thermality of the bath. The non-Markovianity can be greatly enhanced by the non-thermality of the bath.

*Acknowledgement* - S.-W. Li appreciates much for the helpful discussions with H. Dong, A. Svidzinsky, and D. Wang in Texas A&M University. This study is supported by NSF Grant PHY-1241032 and Robert A. Welch Foundation Award A-1261.

## Appendix A: Derivation of the Master equations

Here we show the detailed derivations for the master equation (13) and the time-dependent one Eq. (24). First, through the approximation Markov-2 [Eq. (11)], we have the following equation in the interaction picture,

$$\begin{aligned}\dot{\rho}_S &= -\text{Tr}_B \int_0^\infty ds [H_{SB}(t), [H_{SB}(t-s), \rho_S(t) \otimes \rho_B]] \\ &= \text{Tr}_B \int_0^\infty ds \left\{ H_{SB}(t-s) \rho_S(t) \otimes \rho_B H_{SB}(t) - H_{SB}(t) H_{SB}(t-s) \rho_S(t) \otimes \rho_B \right\} + \text{h.c.}\end{aligned}\quad (\text{A1})$$

Expanding the two terms in the above equation, we obtain (denoting  $\Delta_{mn} := \varepsilon_m - \varepsilon_n$ )

$$\begin{aligned}& \text{Tr}_B \int_0^\infty ds H_{SB}(t-s) \rho_S \otimes \rho_B H_{SB}(t) \\ &= \sum_{m,n=1}^2 \int_0^\infty ds \hat{\tau}_m^+ \rho_S \hat{\tau}_n^- \cdot e^{i\varepsilon_m(t-s)} e^{-i\varepsilon_n t} \cdot \langle \hat{B}_n^\dagger(t) \hat{B}_m(t-s) \rangle + \hat{\tau}_m^- \rho_S \hat{\tau}_n^+ \cdot e^{-i\varepsilon_m(t-s)} e^{i\varepsilon_n t} \cdot \langle \hat{B}_n(t) \hat{B}_m^\dagger(t-s) \rangle \\ &= \sum_{mn} \int_0^\infty ds \int_0^\infty \frac{d\nu}{2\pi} \hat{\tau}_m^+ \rho_S \hat{\tau}_n^- e^{i\Delta_{mn}t} \cdot e^{-i(\varepsilon_m - \nu)s} \cdot J_{nm}(\nu) \tilde{n}(\nu) + \hat{\tau}_m^- \rho_S \hat{\tau}_n^+ e^{-i\Delta_{mn}t} \cdot e^{i(\varepsilon_m - \nu)s} \cdot J_{mn}(\nu) [\tilde{n}(\nu) + 1] \\ &\simeq \sum_{mn} \frac{J_{nm}(\varepsilon_m)}{2} \tilde{n}(\varepsilon_m) \cdot \hat{\tau}_m^+ \rho_S \hat{\tau}_n^- e^{i\Delta_{mn}t} + \frac{J_{mn}(\varepsilon_m)}{2} [\tilde{n}(\varepsilon_m) + 1] \cdot \hat{\tau}_m^- \rho_S \hat{\tau}_n^+ e^{-i\Delta_{mn}t}\end{aligned}\quad (\text{A2})$$



$$\begin{aligned}
& \text{Tr}_B \int_0^\infty ds H_{SB}(t) H_{SB}(t-s) \rho_S(t) \otimes \rho_B \\
&= \sum_{m,n=1}^2 \int_0^\infty ds \hat{\tau}_m^+ \hat{\tau}_n^- \rho_S \cdot e^{i\varepsilon_m t} e^{-i\varepsilon_n(t-s)} \cdot \langle \hat{B}_m(t) \hat{B}_n^\dagger(t-s) \rangle + \hat{\tau}_m^- \hat{\tau}_n^+ \rho_S \cdot e^{-i\varepsilon_m t} e^{i\varepsilon_n(t-s)} \cdot \langle \hat{B}_m^\dagger(t) \hat{B}_n(t-s) \rangle \\
&= \sum_{mn} \int_0^\infty ds \int_0^\infty \frac{d\nu}{2\pi} \hat{\tau}_m^+ \hat{\tau}_n^- \rho_S \cdot e^{i\Delta_{mn}t} e^{i(\varepsilon_n - \nu)s} \cdot J_{nm}(\nu) [\tilde{n}(\nu) + 1] + \hat{\tau}_m^- \hat{\tau}_n^+ \rho_S \cdot e^{-i\Delta_{mn}t} e^{-i(\varepsilon_n - \nu)s} \cdot J_{mn}(\nu) \tilde{n}(\nu) \\
&\simeq \sum_{mn} \frac{J_{nm}(\varepsilon_n)}{2} [\tilde{n}(\varepsilon_n) + 1] \cdot \hat{\tau}_m^+ \hat{\tau}_n^- \rho_S e^{i\Delta_{mn}t} + \frac{J_{mn}(\varepsilon_n)}{2} \tilde{n}(\varepsilon_n) \cdot \hat{\tau}_m^- \hat{\tau}_n^+ \rho_S e^{-i\Delta_{mn}t}
\end{aligned} \tag{A3}$$

In the above calculation, we define the coupling spectrum  $J_{mn}(\omega)$  as

$$J_{mn}(\omega) := 2\pi \sum_k g_{m,k}^* g_{n,k} \delta(\omega - \omega_k) = [J_{nm}(\omega)]^*, \tag{A4}$$

so the bath correlation functions are

$$\begin{aligned}
\langle \hat{B}_n^\dagger(t) \hat{B}_m(t-s) \rangle &= \sum_k g_{n,k}^* g_{m,k} \langle \hat{b}_k^\dagger \hat{b}_k \rangle e^{i\nu s} = \int_0^\infty \frac{d\nu}{2\pi} J_{nm}(\nu) \tilde{n}(\nu) e^{i\nu s}, \\
\langle \hat{B}_n(t) \hat{B}_m^\dagger(t-s) \rangle &= \sum_k g_{m,k}^* g_{n,k} \langle \hat{b}_k \hat{b}_k^\dagger \rangle e^{-i\nu s} = \int_0^\infty \frac{d\nu}{2\pi} J_{mn}(\nu) [\tilde{n}(\nu) + 1] e^{-i\nu s}.
\end{aligned} \tag{A5}$$

Notice that since the non-thermal bath state has the form of Eq. (3), we have  $\text{Tr}[\rho_B \cdot \hat{b}_k^\dagger \hat{b}_q] = \delta_{kq} \cdot \bar{n}_p(\omega_k, T_k)$ , and so we use  $\tilde{n}(\omega_k) := \langle \hat{b}_k^\dagger \hat{b}_k \rangle$  to replace the standard Planck distribution  $\bar{n}_p(\omega, T)$  in thermal baths. For example, a temperature distribution like Eq. (18) would lead to

$$\tilde{n}(\omega) = \begin{cases} \frac{1}{\exp(\omega/T_1) - 1}, & 0 < \omega \leq \bar{\varepsilon} \\ \frac{1}{\exp(\omega/T_2) - 1}, & \omega > \bar{\varepsilon} \end{cases} \tag{A6}$$

In the calculation of Eqs. (A2, A3), we have utilized the formula

$$\int_0^\infty e^{i\omega s} ds = \pi \delta(\omega) + i\mathbf{P} \frac{1}{\omega}, \tag{A7}$$

and the principal integrals in the the imaginary parts are omitted.

Therefore, in the Schrödinger's picture, we obtain

$$\begin{aligned}
\dot{\rho}_S &= i[\rho_S, H_S] + \sum_{m,n=1}^2 \frac{1}{2} J_{nm}(\omega_m) \tilde{n}(\omega_m) \cdot [\hat{\tau}_m^+ \rho_S, \hat{\tau}_n^-] + \text{h.c.} \\
&\quad + \frac{1}{2} J_{mn}(\omega_m) [\tilde{n}(\omega_m) + 1] \cdot [\hat{\tau}_m^- \rho_S, \hat{\tau}_n^+] + \text{h.c.}
\end{aligned} \tag{A8}$$

Defining  $\Gamma_{mn}^+(\omega) := J_{nm}(\omega) \tilde{n}(\omega)$ ,  $\Gamma_{mn}^-(\omega) := J_{mn}(\omega) [\tilde{n}(\omega) + 1]$ , and  $D_{mn}^\pm := [\Gamma_{mn}^\pm(\varepsilon_m) + \Gamma_{mn}^\pm(\varepsilon_n)]/2$ , we can verify that the above master equation can be written in the standard Lindblad form as Eq. (13) with a unitary correction term Eq. (17).

To derive the time-dependent master equation (24), we can change the integral upper limit in Eqs. (A1, A2, A3). Thus, we obtain

$$\begin{aligned}
& \text{Tr}_B \int_0^t ds H_{SB}(t-s) \rho_S \otimes \rho_B H_{SB}(t) \\
&= \sum_{mn} \left[ \int_0^t ds \int_0^\infty \frac{d\nu}{2\pi} e^{-i(\varepsilon_m - \nu)s} J_{nm}(\nu) \tilde{n}(\nu) \right] \hat{\tau}_m^+ \rho_S \hat{\tau}_n^- e^{i\Delta_{mn}t} + \left[ \int_0^t ds \int_0^\infty \frac{d\nu}{2\pi} e^{i(\varepsilon_m - \nu)s} J_{mn}(\nu) [\tilde{n}(\nu) + 1] \right] \hat{\tau}_m^- \rho_S \hat{\tau}_n^+ e^{-i\Delta_{mn}t},
\end{aligned} \tag{A9}$$

$$\begin{aligned}
& \text{Tr}_B \int_0^t ds H_{SB}(t) H_{SB}(t-s) \rho_S(t) \otimes \rho_B \\
&= \sum_{mn} \left[ \int_0^t ds \int_0^\infty \frac{d\nu}{2\pi} e^{i(\varepsilon_n - \nu)s} J_{nm}(\nu) [\tilde{n}(\nu) + 1] \right] \hat{\tau}_m^+ \hat{\tau}_n^- \rho_S e^{i\Delta_{mn}t} + \left[ \int_0^t ds \int_0^\infty \frac{d\nu}{2\pi} e^{-i(\varepsilon_n - \nu)s} J_{mn}(\nu) \tilde{n}(\nu) \right] \hat{\tau}_m^- \hat{\tau}_n^+ \rho_S e^{-i\Delta_{mn}t}.
\end{aligned} \tag{A10}$$

The coefficients become time-dependent, so we define the decay rates as [Eq. (25)]

$$\begin{aligned}\Gamma_{mn}^+(\omega, t) &:= \Re \int_0^t ds \int_0^\infty \frac{d\nu}{2\pi} J_{nm}(\nu) \tilde{n}(\nu) e^{i(\nu-\omega)s}, \\ \Gamma_{mn}^-(\omega, t) &:= \Re \int_0^t ds \int_0^\infty \frac{d\nu}{2\pi} J_{mn}(\nu) [\tilde{n}(\nu) + 1] e^{-i(\nu-\omega)s}.\end{aligned}\quad (\text{A11})$$

Here we also omit the imaginary part, so  $\Gamma_{mn}^\pm(\omega, t)$  will return to the homogenous Markovian case directly when  $t \rightarrow \infty$ . With these changes, the time-dependent master equation is

$$\begin{aligned}\dot{\rho}_S &= i[\rho_S, H_S] + \sum_{m,n=1}^2 \frac{1}{2} \Gamma_{mn}^+(\varepsilon_m, t) \cdot [\hat{\tau}_m^+ \rho_S, \hat{\tau}_n^-] + \text{h.c.} \\ &\quad + \frac{1}{2} \Gamma_{mn}^-(\varepsilon_m, t) \cdot [\hat{\tau}_m^- \rho_S, \hat{\tau}_n^+] + \text{h.c.}\end{aligned}\quad (\text{A12})$$

which is similar to Eq. (A8). We can verify that this master equation can be also written in the form of Eq. (24).

- 
- [1] L.-M. Duan and G.-C. Guo, *Phys. Rev. Lett.* **79**, 1953 (1997).
  - [2] P. Zanardi and M. Rasetti, *Phys. Rev. Lett.* **79**, 3306 (1997).
  - [3] D. A. Lidar, I. L. Chuang, and K. B. Whaley, *Phys. Rev. Lett.* **81**, 2594 (1998).
  - [4] M. O. Scully and M. S. Zubairy, *Quantum optics* (Cambridge university press, 1997).
  - [5] J. Roßnagel, O. Abah, F. Schmidt-Kaler, K. Singer, and E. Lutz, *Phys. Rev. Lett.* **112**, 030602 (2014).
  - [6] R. Alicki and D. Gelbwaser-Klimovsky, *New J. Phys.* **17**, 115012 (2015).
  - [7] K. E. Dorfman, D. V. Voronine, S. Mukamel, and M. O. Scully, *Proc. Nat. Acad. Sci.* **110**, 2746 (2013).
  - [8] C. Creatore, M. A. Parker, S. Emmott, and A. W. Chin, *Phys. Rev. Lett.* **111**, 253601 (2013).
  - [9] J. Olšina, A. G. Dijkstra, C. Wang, and J. Cao, *arXiv:1408.5385* (2014).
  - [10] B. L. Hu, J. P. Paz, and Y. Zhang, *Phys. Rev. D* **45**, 2843 (1992).
  - [11] H. Breuer and F. Petruccione, *The theory of open quantum systems* (Oxford University Press, 2002).
  - [12] J. Ma, Z. Sun, X. Wang, and F. Nori, *Phys. Rev. A* **85**, 062323 (2012).
  - [13] L.-P. Yang, C. Y. Cai, D. Z. Xu, W.-M. Zhang, and C. P. Sun, *Phys. Rev. A* **87**, 012110 (2013).
  - [14] Z.-Z. Li, C.-T. Yip, H.-Y. Deng, M. Chen, T. Yu, J. Q. You, and C.-H. Lam, *Phys. Rev. A* **90**, 022122 (2014).
  - [15] U. Weiss, *Quantum dissipative systems* (World Scientific, 2012).
  - [16] G. S. Agarwal, *Quantum statistical theories of spontaneous emission and their relation to other approaches* (Springer, 1974).
  - [17] S.-Y. Zhu, L. M. Narducci, and M. O. Scully, *Phys. Rev. A* **52**, 4791 (1995).
  - [18] Z.-H. Li, D.-W. Wang, H. Zheng, S.-Y. Zhu, and M. S. Zubairy, *Phys. Rev. A* **82**, 050501 (2010).
  - [19] D. Xu, C. Wang, Y. Zhao, and J. Cao, *New J. Phys.* **18**, 023003 (2016).
  - [20] M. Orszag, *Quantum optics* (Springer, 2000).
  - [21] S.-W. Li, L.-P. Yang, and C.-P. Sun, *Eur. Phys. J. D* **68**, 45 (2014), *arXiv:1303.1266*.
  - [22] S.-W. Li, C. Y. Cai, and C. P. Sun, *Ann. Phys.* **360**, 19 (2015).
  - [23] W. W.-S. Wei, *Time series analysis* (Addison-Wesley Redwood City, California, 1994).
  - [24] H. Wichterich, M. J. Henrich, H.-P. Breuer, J. Gemmer, and M. Michel, *Phys. Rev. E* **76**, 031115 (2007).
  - [25] P. G. Bergmann and J. L. Lebowitz, *Phys. Rev.* **99**, 578 (1955).
  - [26] C.-Y. Cai, S.-W. Li, X.-F. Liu, and C. P. Sun, *arXiv:1407.2004* (2014).
  - [27] A. Suárez, R. Silbey, and I. Oppenheim, *J. Chem. Phys.* **97**, 5101 (1992).
  - [28] V. Gorini, A. Kossakowski, and E. C. G. Sudarshan, *J. Math. Phys.* **17**, 821 (1976).
  - [29] G. Lindblad, *Comm. Math. Phys.* **48**, 119 (1976).
  - [30] A. A. Clerk, M. H. Devoret, S. M. Girvin, F. Marquardt, and R. J. Schoelkopf, *Rev. Mod. Phys.* **82**, 1155 (2010).
  - [31] C. Gardiner and P. Zoller, *Quantum noise*, Vol. 56 (Springer, 2004).
  - [32] Á. Rivas, S. F. Huelga, and M. B. Plenio, *Rep. Prog. Phys.* **77**, 094001 (2014).
  - [33] D. Chruściński and A. Kossakowski, *J. Phys. B* **45**, 154002 (2012).
  - [34] M. J. W. Hall, *J. Phys. A* **41**, 205302 (2008).